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(54) Process for preparing poly (trimethylene terephthalate) yarns

(57) Poly(trimethylene terephthalate) is formed into a bulk continuous filament yarn by a process comprising melt-spinning poly(trimethylene terephthalate) to pro-

duce a plurality of spun filaments; forming the spun filaments into a yarn; and drawing the yarn in a two-stage drawing process wherein the second stage draw is at a significantly higher draw ratio than the first draw ratio.

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Description

This invention relates to the spinning of poly(trimethylene terephthalate) into yarn suitable for carpets.

Polyesters prepared by condensation polymerization of the reaction product of a diol with a dicarboxylic acid can be spun into yarn suitable for carpet fabric. U.S. 3,998,042 describes a process for preparing poly(ethylene terephthalate) yarn in which the extruded fiber is drawn at high temperature (160°C) with a steam jet assist, or at a lower temperature (95°C) with a hot water assist. Poly(ethylene terephthalate) can be spun into bulk continuous filament (BCF) yarn in a two-stage drawing process in which the first stage draw is at a significantly higher draw ratio than the second stage draw. U.S. 4,877,572 describes a process for preparing poly(butylene terephthalate) BCF yarn in which the extruded fiber is drawn in one stage, the feed roller being heated to a temperature 30°C above or below the T_g of the polymer and the draw roller being at least 100°C higher than the feed roll. However, the application of conventional polyester spinning processes to prepare poly(trimethylene terephthalate) BCF results in yarn which is of low quality and poor consistency.

It has now been found that poly(trimethylene) terephthalate can be melt-spun into high quality BCF yarn by using a two-stage drawing process in which the second stage draw is at a significantly higher draw ratio than the first stage. The present invention therefore provides a process for preparing bulk continuous fiber yarn from poly(trimethylene terephthalate) comprising:

- (a) melt-spinning poly(trimethylene terephthalate), suitably at a temperature within the range of 250 to 280°C, to produce a plurality of spun filaments;
- (b) cooling the spun filaments;
- (c) converging the spun filaments into a yarn;
- (d) drawing the yarn at a first draw ratio within the range of 1.05 to 2 in a first drawing stage defined by at least one feed roller and at least one first draw roller, each feed roller being heated to a temperature less than 100°C and each draw roller being heated to a temperature greater than the temperature of said feed roller and within the range of 80 to 150°C;
- (e) subsequently drawing the yarn at a second draw ratio of at least 2.2 times that of the first draw ratio in a second drawing stage defined by said first draw roller and at least one second draw roller, each second draw roller being heated to a temperature greater than said first draw roller and within the range of 100 to 200°C; and
- (f) winding the drawn yarn.

The process may optionally include texturing the drawn yarn prior to or after winding step (f).

The fiber-spinning process is designed specifically for poly(trimethylene terephthalate), the product of the condensation polymerization of the reaction product of trimethylene diol (also called "1,3-propane diol") and a terephthalic acid or an ester thereof, such as terephthalic acid and dimethyl terephthalate. The poly(trimethylene terephthalate) may also include minor amounts of the derivatives of other monomers such as ethane diol and butane diol as well as minor amounts of the derivatives of other diacids or diesters such as isophthalic acid. Poly(trimethylene terephthalate) having an intrinsic viscosity (i.v.) within the range of 0.8 to 1.0 dl/g, preferably 0.86 to 0.96 dl/g (as measured in a 50/50 mixture of methylene chloride and trifluoroacetic acid at 30°C) and a melting point within the range of 215 to 230°C is particularly suitable. The moisture content of the poly(trimethylene terephthalate) should be less than 0.005% prior to extrusion. Such a moisture level can be achieved by, for example, drying polymer pellets in a dryer at 150-180°C until the desired dryness has been achieved.

One embodiment of the invention process can be described by reference to Figure 1. Molten poly(trimethylene terephthalate) which has been extruded through a spinneret into a plurality of continuous filaments 1 at a temperature within the range of 240 to 280°C, preferably 250 to 270°C, and then cooled rapidly, preferably by contact with cold air, is converged into a multifilament yarn and the yarn is passed in contact with a spin finish applicator, shown here as kiss roll 2. Yarn 3 is passed around denier control rolls 4 and 5 and then to a first drawing stage defined by feed roll 7 and draw roll 9. Between rolls 7 and 9, yarn 8 is drawn at a relatively low draw ratio, within the range of 1.05 to 2, preferably 1.10 to 1.35. Roller 7 is maintained at a temperature less than about 100°C, preferably within the range of 40 to 85°C. Roller 9 is maintained at a temperature within the range of 80 to 150°C, preferably 90 to 140°C. Drawn yarn 10 is passed to a second drawing stage, defined by draw rolls 9 and 11. The second-stage draw is carried out at a draw ratio at least 2.2 times that of the first stage draw ratio, preferably at a draw ratio within the range of 2.2 to 3.4 times that of the first stage. Roller 11 is maintained at a temperature within the range of 100 to 200°C. In general, the three rollers will be sequentially higher in temperature. The selected temperature will depend upon other process variables, such as whether the BCF is made with separate drawing and texturing steps or in a combined draw/texturing process, the effective heat transfer of the rolls used, residence time on the roll, and whether second heated roll upstream of the texturing jet. Drawn fiber 12 is passed in contact with optional relaxation stabilization of the drawn yarn. Stabilized yarn 14 is passed to optional winder 15 or is sent directly to the next step.

process.

The drawn yarn is bulked by suitable means such as a hot air texturing jet. The preferred feed roll temperature for texturing is within the range of 150 to 200°C. The texturing air jet temperature is generally within the range of 150 to 210°C, and the texturing jet pressure is generally within the range of 340 to 825 kPa to provide a high-bulk BCF yarn. Wet or superheated steam can be substituted for hot air as the bulking medium.

Figure 2 shows an embodiment of the two-stage drawing process which includes texturing steps downstream of the drawing zone. Molten poly(trimethylene terephthalate) is extruded through spinneret 21 into a plurality of continuous filaments 22 and is then quenched by, for example, contact with cold air. The filaments are converged into yarn 24 to which spin finish is applied at 23. Yarn 27 is advanced to the two-stage draw zone via non-heated rolls 25 and 26.

In the first draw stage, yarn 31 is drawn between feed roll 28 and draw roll 29 at a draw ratio within the range of 1.05 and 2. Drawn yarn 32 is then subjected to a second draw at a draw ratio at least 2.2 times the first draw ratio, preferably a draw ratio within the range of 2.2 to 3.4 times that of the first draw. The temperature of roll 28 is less than 100°C. The temperature of draw roll 29 is within the range of 80 to 150°C. The temperature of draw roll 30 is within the range of 100 to 200°C. Drawn yarn 33 is advanced to heated rolls 34 and 35 to preheat the yarn for texturing. Yarn 36 is passed through texturing air jet 37 for bulk enhancement and then to jet screen cooling drum 38. Textured yarn 39 is passed through tension control 40, 41 and 42 and then via idler 43 to optional entangler 44 for yarn entanglement if desired for better processing downstream. Entangled yarn 45 is then advanced via idler 46 to an optional spin finish applicator 47 and is then wound onto winder 48. The yarn can then be processed by twisting, texturing and heat-setting as desired and tufted into carpet as is known in the art of synthetic carpet manufacture.

Poly(trimethylene terephthalate) yarn prepared by the invention process has high bulk (generally within the range of 20 to 45%, preferably within the range of 26 to 35%), resilience and elastic recovery, and is useful in the manufacture of carpet, including cut-pile, loop-pile and combination-type carpets, mats and rugs. Poly(trimethylene terephthalate) carpet has been found to exhibit good resiliency, stain resistance and dyability with disperse dyes at atmospheric boil with optional carrier.

Example 1

Effect of Intrinsic Viscosity on Poly(trimethylene terephthalate) Fiber Drawing

Four poly(trimethylene terephthalate) polymers having intrinsic viscosities of 0.69, 0.76, 0.84 and 0.88 dL/g, respectively, were each spun into 70 filaments with trilobal cross-sections using a spinning machine having a take-up and drawing configuration as shown in Figure 1. Roll 1 (see detail below) was a double denier control roll; roll 2 ran at a slightly higher speed to maintain a tension and act as a feed roll for drawing. First stage drawing took place between rolls 2 and 3, and second-stage drawing took place between rolls 3 and 4. The drawn yarn contacted relax roll 5 prior to wind-up. The spin finish was a 15% Lurol PF 4358-15 solution from G.A. Goulston Company applied with a kiss roll. Fiber extrusion and drawing conditions for each polymer were as follows:

Extrusion Conditions			
Polymer IV (dL/g):		0.84, 0.88	0.69, 0.76
	Units		
Extruder Temp. Profile:			
Zone 1	°C	230	225
Zone 2	°C	250	235
Zone 3	°C	250	235
Zone 4	°C	250	235
Melt Temp.	°C	255	240
Extrusion Pack Pressure	kPa	12710-19700	3500-9000
Denier Control Roll Speed	m/min.	225	220

Fiber Drawing Conditions				
Polymer IV (dl/g)	0.88	0.84	00.76	0.69
Roll Temp.: °C				
Roll 2	80	80	80	80
Roll 3	95	95	95	95
Roll 4	155	155	155	155
Roll 5	RT	RT	RT	RT
Roll Speeds: m/min.				
Roll 2	230	230	230	230
Roll 3	310	310	404	404

Fiber Drawing Conditions (continued)				
Roll 4	1020	1165	1089	1089
Roll 5	1035	1102	1075	1075
First Stage Draw Ratio	1.35	1.35	1.76	1.76
Second Stage Draw Ratio	3.29	3.29	2.70	2.70

Fiber tensile properties are shown in Table 1.

TABLE 1

Run	I.V. (dl/g)	Yarn Count (den.)	Tenacity (g/den.)	% Elongation
1	0.69	1182	1.51	70.7
2	0.76	1146	1.59	79.7
3	0.84	1167	2.03	89.0
4	0.88	1198	2.24	67.5

Poly(trimethylene terephthalate) of intrinsic viscosities 0.69 and 0.76 (Runs 1 and 2) have a second stage draw ratio only 1.53 greater than that of the first stage draw ratio, i.e. below the 2.2 minimum ratio of the present invention, and are included for comparative purpose. These comparative runs gave yarn of inferior tensile properties compared with the yarn of Runs 3 and 4 (which illustrate the invention). These polymers were re-spun at a lower extruder temperature profile. Although they could be spun and drawn, the fibers had high die swell. When the fiber cross-sections were examined with an optical microscope, the 0.69 i.v. fibers swelled to a point that they were no longer trilobal in shape and resembled delta cross-sections. They also had relatively low tenacity.

Example 2Two-Stage Drawing of PTT Fibers

0.68 l.v. poly(trimethylene terephthalate) was extruded into 72 filaments having trilobal cross-section using a fiber-spinning machine having take-up and drawing configurations as in Example 1. Spin finish was applied as in Example 1. Extrusion and drawing conditions were as follows.

Extrusion Conditions		
Extruder Temperature Profile:	Units	
Zone 1	°C	230
Zone 2	°C	260
Zone 3	°C	260
Zone 4	°C	260
Melt Temp.	°C	265
Denier Control Roll Speed	m/min.	230

Fiber Drawing Conditions

Runs	5	6	7	8	9	10	11
	Units						
Roll 2 Temp./Speed	80/235	80/235	100/235	100/235	100/235	100/235	100/235
Roll 3 Temp./Speed	90/317	100/286	100/817	100/817	100/817	100/993	100/945
Roll 4 Temp./Speed	155/1123	100/1021	155/1047	140/1103	140/1145	130/1044	140/996
Roll 5 Temp./Speed	RT/1096	RT/1011	RT/1029	RT/1082	RT/1134	RT/1019	RT/981
1st Stage Draw Ratio	1.35	1.22	3.48	3.48	3.48	4.23	4.02
2nd Stage Draw Ratio	3.55	3.57	1.28	1.35	1.40	1.05	1.05
Total Draw Ratio	4.79	4.36	4.45	4.70	4.87	4.44	4.22
Yarn Count, den.	1225	1281	1275	1185		1210	1288
Tenacity, g/den.	1.95	1.95	1.61	1.32		1.85	1.11
Elongation	55	75	70	76		78	86

It was observed during spinning and drawing that, when the first-stage draw ratio (between rolls 2 and 3) was less than about 1.5, and the second stage draw ratio was 2.63 greater than that of the first stage draw ratio (i.e. in conformity with the present invention), as in Runs 5 and 6, there were fewer broken filaments and the tenacities of the filaments were generally higher than when first-stage draw was higher than 1.5. When the first-stage draw was increased to greater than 3 and the second stage draw ratio was less than that of the first stage (i.e. illustrative of prior art spinning processes, and therefore included for comparative purposes; Runs 7, 8, 9, 10, and 11), it was observed that the fibers had a white streaky appearance, the threadlines were loopy, and there were frequent filament wraps on the draw rolls. The process was frequently interrupted with fiber breaks.

Example 3

Spinning, Drawing and Texturing Poly(trimethylene terephthalate) BCF to High Bulk

The extrusion conditions in this experiment were the same as in Example 2. The fibers were spun, drawn and wound as in Example 1. They were then textured by heating the fibers on a feed roll and exposing the fibers to a hot air jet. The textured fibers were collected as a continuous plug on a jet-screen cooling drum. Partial vacuum was applied to the drum to pull the ambient air to cool the yarns and keep them on the drum until they were wound. The yarns were air entangled between the drum and the winder. The feed roll and texturizer air jet temperatures were kept constant, and the air jet pressure was varied from 350 to 700 kPa to prepare poly(trimethylene terephthalate) BCF of various bulk levels.

Drawing and texturing conditions were as follows.

Drawing Conditions		
Rolls	Temperature, C	Speed, m/min.
Roll 1	RT	225
Roll 2	80	230
Roll 3	95	264
Roll 4	90	1058
Roll 5	110	1042

Texturing Conditions	
Feed Roll Temperature, °C	180
Feed Roll Speed, m/min.	980
Air Jet Temperature, °C	180
Interlacing Pressure, kPa	70

Yarn bulk and shrinkage were measured by taking 18 wraps of the textured yarn in a denier creel and tying it into a skein. The initial length L_0 of the skein was 560 mm in English unit creel. A 1g weight was attached to the skein and it was hung in a hot-air oven at 130°C for 5 minutes. The skein was removed and allowed to cool for 3 minutes. A 50g weight was then attached and the length L_1 was measured after 30 seconds. The 50g weight was removed, a 4.5 kg weight was attached, and the length L_2 was measured after 30 seconds. Percent bulk was calculated as $(L_0 - L_1)/L_0 \times 100\%$ and shrinkage was calculated as $(L_0 - L_2)/L_0 \times 100\%$. Results are shown in Table 2.

TABLE 2

Package No.	Yarn Count, den.	% Bulk	% Shrinkage
T50	1437	32.6	3.6
T60	1406	35.7	2.7
T70	1455	39.4	3.2
T80	1500	38.0	3.6

TABLE 2 (continued)

Package No.	Yarn Count, den.	% Bulk	% Shrinkage
T90	1525	37.6	4.1
T100	1507	38.0	3.8

The experiment showed that poly(trimethylene terephthalate) BCF can be textured to high bulk with a hot air texturizer.

Example 4

Carpet Resiliency Comparison

Poly(trimethylene terephthalate) BCF yarns were made in two separate steps: (1) spinning and drawing set-up as in Example 1 and (2) texturing. Extrusion, drawing and texturing conditions for the poly(trimethylene terephthalate) yarns were as follows.

Extrusion Conditions		
Extruder Temperature	Units	
Zone 1	°C	240
Zone 2	°C	255
Zone 3	°C	255
Zone 4	°C	255
Melt Temperature	°C	260
Pack Pressure	kPa	12800

Drawing Conditions		
	Units	
Roll 1 Temp./Speed	°C/m/min.	RT/223
Roll 2 Temp./Speed	°C/m/min.	80/230
Roll 3 Temp./Speed	°C/m/min.	95/288
Roll 4 Temp./Speed	°C/m/min.	150/1088
Roll 5 Temp./Speed	°C/m/min.	TY/1000

Texturing Conditions		
	Units	
Feed Roll Temp.	°C	180
Feed Roll Speed	m/min.	980
Air Jet Temp.	°C	180
Air Jet Pressure	kPa	630
Interlacing Pressure	kPa	70

The yarn produced was 1150 denier with 2.55 g/den tenacity and 63% elongation. The textured yarn was twisted, heat set as indicated, and tufted into carpets. Performances of the poly(trimethylene terephthalate) carpets were compared with a commercial 1100 denier nylon 66 yarn. Results are shown in Table 3.

TABLE 3

Run	Twist/25.4 mm	Heat Setting Conditions	Accelerated Floor Traffic	% Loss in Pile Thickness
12 (Poly(trimethylene terephthalate))	4.5 x 4.5	133°C Autoclave	3.75	2.4
13 (Poly(trimethylene terephthalate))	4.5 x 4.5	180°C Seussen	3.5	7.1
14 (Poly(trimethylene terephthalate))	5.0 x 5.0	133°C Autoclave	3.75	1.7
15 nylon 66	4.0 x 4.0	133°C Autoclave	3.0	6.4
16 nylon 66	4.0 x 4.0	190°C Seussen	3.5	4.5

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The heat-set yarns were tufted into 680 g cut-pile Saxony carpets in 3.2 mm gauge, 14.3 mm pile height, and dyed with disperse blue 56 (without a carrier) at atmospheric boil into medium blue color carpets. Visual inspection of the finished carpets disclosed that the poly(trimethylene terephthalate) carpets (Runs 12, 13 and 14) had high bulk and excellent coverage which were equal to or better than the nylon controls (Runs 15 and 16). Carpet resiliency was tested in accelerated floor trafficking with 20,000 footsteps. The appearance retention was rated 1 (severe change in appearance), 2 (significant change), 3 (moderate change), 4 (slight change) and 5 (no change). As can be seen in Table 3, the poly(trimethylene terephthalate) carpets were equal to or better than the nylon 66 controls in the accelerated walk tests and in percent thickness loss.

Example 5

One-Step Processing of Poly(trimethylene terephthalate) BCF Yarn from Spinning to Texturing

Poly(trimethylene terephthalate) (I.v. 0.90) was extruded into 72 trilobal cross-section filaments. The filaments were processed on a line as shown in Figure 2 having two cold rolls, three draw rolls and double yarn feed rolls prior to texturing. The yarns were textured with hot air, cooled in a rotating jet screen drum and wound up with a winder. Luroi NF 3278 CS (G.A. Goulston Co.) was used as the spin finish. Texturing conditions were varied to make poly(trimethylene terephthalate) BCF yarns having different bulk levels. Extrusion, drawing, texturing and winding conditions were as follows.

Extrusion Conditions		
Extruder Temperature Profiles	Units	
Zone 1	°C	240
Zone 2	°C	260
Zone 3	°C	260
Zone 4	°C	265
Melt Temperature	°C	265
Pump Pressure	kPa	25500

Drawing Conditions		
	Temperature °C	Speed, m/min.
Cold Roll 1	RT	211
Cold Roll 2	RT	264
Draw Roll 1	50	290
Draw Roll 2	90	330
Draw Roll 3	110	1100

The yarns were twisted, heat set and tufted into carpets for performance evaluation. Results are shown in Table 4.

TABLE 4

Sample Number	Feed Roll Temp, °C	Texturizing Jet Temp., °C	Texturizing Press., kpa	Yarn count den.	% Bulk	% Shrinkage	Accelerated Walk Test Rating
1	150	180	500	1490	19.2	1.58	3.25
2	150	180	770	1420	26	1.59	3.5
3	150	200	770	1546	30.5	1.59	3.0
4	180	180	500	1429	24.6	2.04	3.0
5	180	180	770	1496	29.8	1.81	3.5
6	180	200	500	1475	26.5	1.36	2.75
7	180	200	770	1554	32.8	0.86	3.0
8	150	190	630	1482	26	2.31	3.25
9	180	190	630	1430	29	1.58	3.5
10	165	190	630	1553	29	2.26	3.75
Nylon 6							3.5
Nylon 66							3.5

Example 6**Effects of Draw Ratio and Roll Temperature on Yarn Properties**

Poly(trimethylene terephthalate) (0.90 i.v.) was spun into 72 filaments with trilobal cross-sections using a machine as described in Example 5. Extrusion conditions were as follows.

Extrusion Conditions		
Extruder Temperature Profiles	Units	
Zone 1	°C	240
Zone 2	°C	260
Zone 3	°C	260
Zone 4	°C	260
Melt Temperature	°C	260

The poly(trimethylene terephthalate) BCF yarns and commercial nylon 6 and 66 yarns were tufted into 900 g. 5/32 gauge cut-pile Saxony carpets having 16 mm pile height. They were walk-tested with 20,000 footsteps accelerated floor trafficking for resiliency and appearance retention comparisons. Roll conditions and results are shown in Table 5.

TABLE 5

Sample:	1	2	3	4	5	nylon 6	nylon 66
Roll 1 Temp.	50	50	50	50	50		
Roll 2 Temp.	90	90	90	90	90		
Roll 3 Temp.	110	110	110	150	150		
Roll 1 Speed	290	290	290	290	290		
Roll 2 Speed	330	330	330	330	330		
Roll 3 Speed	1000	1100	1150	1100	1000		
Draw Ratio	3.45	3.79	3.97	3.97	3.45		
Feed Roll Temp.	165	165	165	165	165		
Feed Roll Speed	1000	1100	1150	1100	1000		
Texturing Jet Temp.	190	190	190	190	190		
Texturing Jet Pressure	630	630	630	630	630		
Interlacing Pressure	210	210	210	210	210		
Bulk	26.1	31.6	31.9	35.8	33		
Shrinkage	1.75	2.04	2.13	2.26	1.92		
Walk Test Rating	4.0	3.5	3.5	3.5	3.5	3.5	3.5

Claims

1. A process for preparing bulk continuous fiber yarn from poly(trimethylene terephthalate) comprising:
 - (a) melt-spinning poly(trimethylene terephthalate) to produce a plurality of spun filaments;
 - (b) cooling the spun filaments;
 - (c) converging the spun filaments into a yarn;
 - (d) drawing the yarn at a first draw ratio within the range of 1.05 to 2 in a first drawing stage defined by at least one feed roller and at least one first draw roller, each feed roller being heated to a temperature less than 100°C and each draw roller being heated to a temperature greater than the temperature of said feed roller and within the range of 80 to 150°C;
 - (e) subsequently drawing the yarn at a second draw ratio of at least 2.2 times that of the first draw ratio in a second drawing stage defined by (the last of) said first draw roller(s) and at least one second draw roller, each second draw roller being heated to a temperature greater than said (last) first draw roller and within the range of 100 to 200°C; and
 - (f) winding the drawn yarn.
2. The process as claimed in claim 1 in which each feed roller is heated to a temperature within the range of 40 to 85°C.
3. The process as claimed in claim 1 or 2 in which the first draw ratio is within the range of 1.10 to 1.35.
4. The process as claimed in claim 1, 2 or 3 in which the second draw ratio is within the range of 2.2 to 3.4 times the first draw ratio.
5. The process as claimed in any one of the preceding claims in which the poly(trimethylene terephthalate) has an intrinsic viscosity within the range of about 0.80 to about 1.0 dl/g.
6. Process as claimed in any one of the preceding claims wherein the drawn yarn is submitted to a texturing treatment.
7. The process as claimed in claim 6 in which texturing is carried out with an air jet at a pressure within the range of 340 to 825 kPa.
8. The process as claimed in claim 6 or 7 in which the texturing step is carried out at a temperature within the range of 150 to 210°C.
9. A carpet the fibers of which consist essentially of poly(trimethylene terephthalate) yarn having a bulk greater than 20 percent and prepared by a process as claimed in any one of the preceding claims.

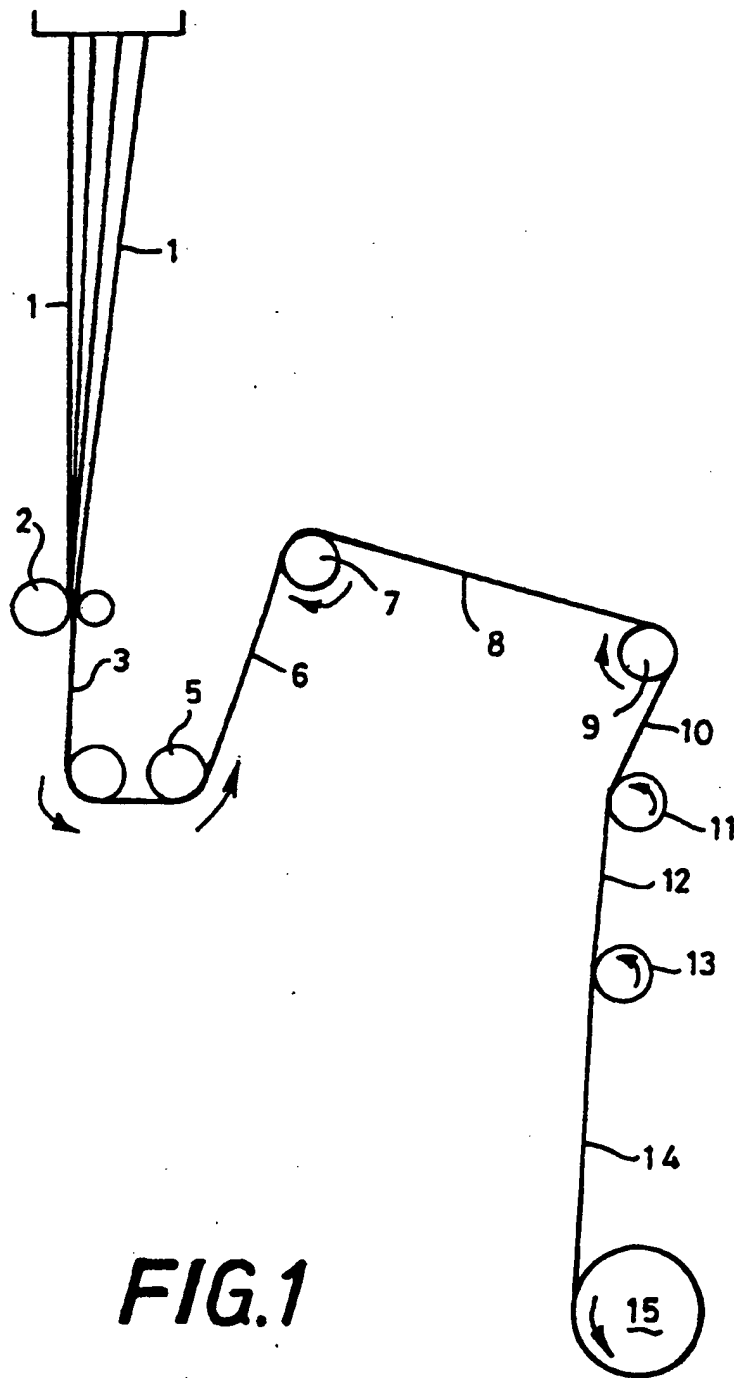


FIG.1

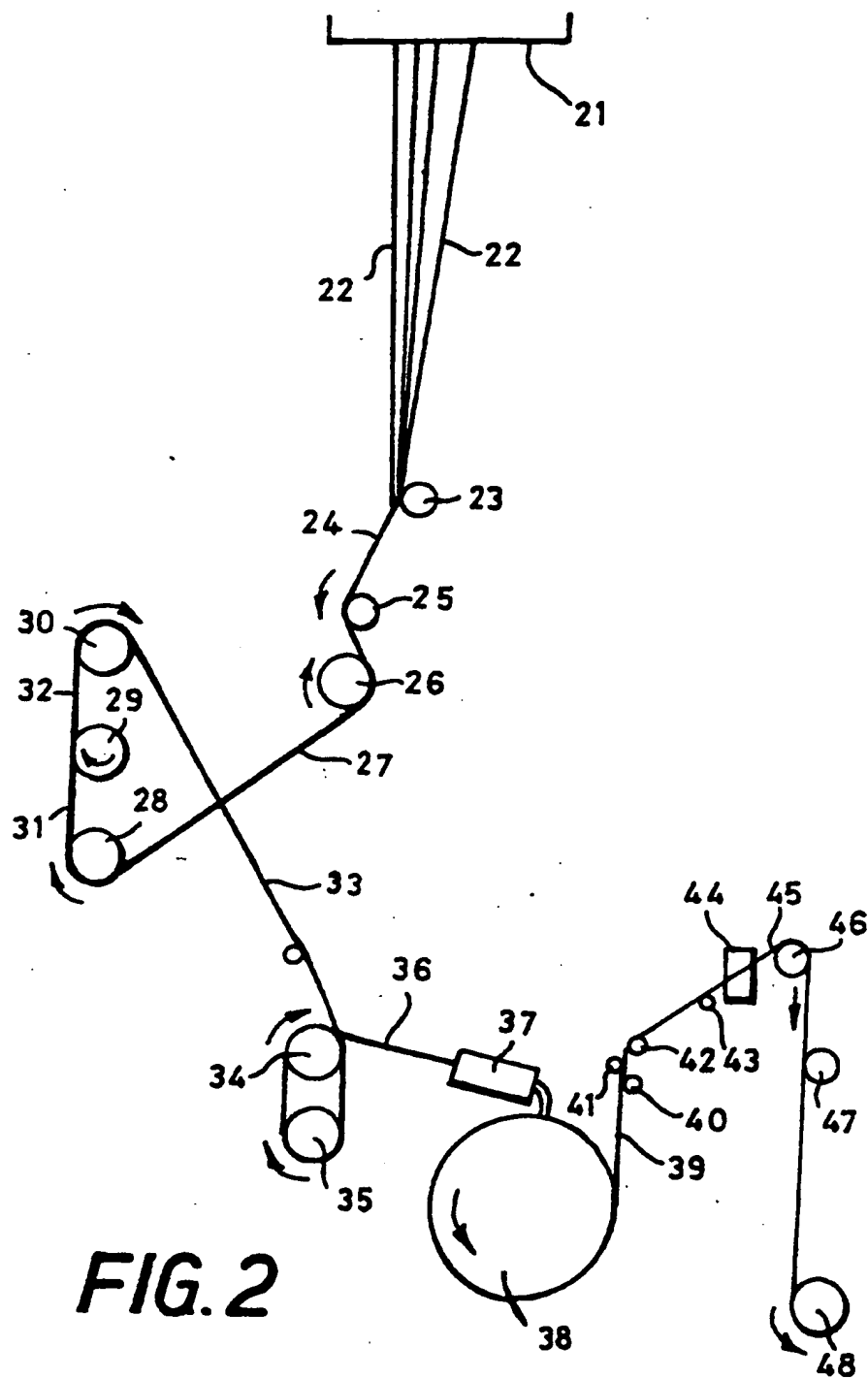


FIG. 2



European Patent
Office

EUROPEAN SEARCH REPORT

Application Number
EP 96 20 1241

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
A	DATABASE WPI Section Ch, Week 8330 Derwent Publications Ltd., London, GB; Class A23, AN 83-720837 XP00014077 & JP-A-58 104 216 (TEIJIN), 21 June 1983 * abstract *	1	D02J1/22 D01F6/62
A	EP-A-0 052 845 (HOECHST) * page 3, line 36 - page 4, line 4; claims 1,4 *	1	
A	WO-A-91 09999 (E.I. DU PONT DE NEMOURS) * page 7, line 34 - page 9, line 30 * * page 14, line 5 - line 25 *	1	
P,A	WO-A-96 00808 (E.I. DU PONT DE NEMOURS) * abstract *	1	
			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
			D02J D01F
The present search report has been drawn up for all claims			
Place of search		Date of completion of the search	Searcher
THE HAGUE		23 September 1996	Goovaerts, R
<p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons A : member of the same patent family, corresponding document</p>			

EPF FORM 1500 (04/94) (PUBLISHED)